



AUG 04 2005

Practitioner's Docket No.: 791_130 RCE

PATENT

ON APPEAL

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE BEFORE THE
HONORABLE BOARD OF PATENT APPEALS AND INTERFERENCES**

In re the application of: Li YANG and Toshihiro YOSHIDA

Ser. No.: 09/770,725

Group Art Unit: 1746

Filed: January 26, 2001

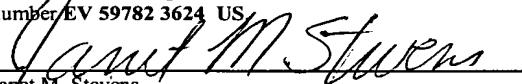
Examiner: Jonathan Crepeau

Confirmation No.: 6015

For: LITHIUM SECONDARY BATTERY

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Janet M. Stevens

TRANSMITTAL OF BRIEF ON APPEAL

Sir:

Transmitted herewith are three copies of the Appellants Brief on Appeal together with a check in the amount of \$500.00 to cover the fee under 37 CFR 1.17(c).

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,

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Enclosures: Brief on Appeal (3)

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Janet M. Stevens
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**TRANSMITTAL OF APPEAL BRIEF
(PATENT APPLICATION--37 C.F.R. § 1.192)**

1. Transmitted herewith, in triplicate, is the APPEAL BRIEF in this application, with respect to the Notice of Appeal filed on June 14, 2005.

2. STATUS OF APPLICANT

This application is on behalf of other than a small entity.
 a small entity.

3. FEE FOR FILING APPEAL BRIEF

Pursuant to 37 C.F.R. § 1.17(c), the fee for filing the Appeal Brief is:

small entity \$250.00
 other than a small entity \$500.00

Appeal Brief Fee Due **\$500.00**

4. EXTENSION OF TERM

The proceedings herein are for a patent application and the provisions of 37 C.F.R. § 1.136 apply.

(complete (a) or (b), as applicable)

(a) [] Applicant petitions for an extension of time under 37 C.F.R. § 1.136 (fees: 37 C.F.R. §1.17(a)(1)-(5)) for the total number of months checked below:

Extension (months)	Fee for other than small entity	Fee for small entity
one month	\$ 120.00	\$ 60.00
two months	\$ 450.00	\$225.00
three months	\$ 1,020.00	\$510.00
four months	\$1,590.00	\$795.00
	Fee	\$.00

(b) [X] Applicant believes that no extension of term is required. However, this conditional petition is being made to provide for the possibility that applicant has inadvertently overlooked the need for a petition and fee for extension of time.

5. TOTAL FEE DUE

The total fee due is: Appeal brief fee \$ 500.00
 Extension fee (if any) \$ _____

TOTAL FEE DUE \$ 500.00

6. FEE PAYMENT

Attached is a check in the sum of \$500.00.
 Charge Account No. _____ the sum of \$ _____.

A duplicate of this transmittal is attached.

7. FEE DEFICIENCY

If any additional extension and/or fee is required, this is a request therefor and to charge Account No. 50-1446.

AND/OR

If any additional fee for claims is required, charge Account No. 50-1446.

Respectfully submitted,

August 4, 2005

Date

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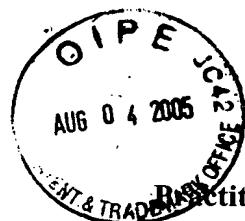
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BRIEF ON APPEAL

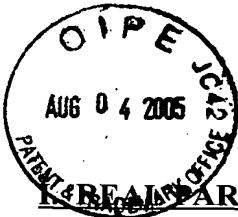
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August 4, 2005



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REAL PARTY IN INTEREST

The real party in interest in the present appeal is NGK Insulators, Ltd., as reflected by an Assignment document recorded in the USPTO microfilm at reel 011490, frames 0426-0428.

II. RELATED APPEALS AND INTERFERENCES

There are no appeals or interferences regarding any application which is related to the present application.

III. STATUS OF CLAIMS

Claims 1-17 (set forth in Appendix A attached hereto) are pending. Claims 1-17 stand finally rejected.

IV. STATUS OF AMENDMENTS

No Amendment has been filed in this application after the presently-appealed final rejection (dated January 14, 2005).

V. SUMMARY OF CLAIMED SUBJECT MATTER

It is respectfully noted that the following concise explanation of the subject matter defined in each of the independent claims involved in the present appeal is provided only to comply with 37 C.F.R. 41.37, and that in order to ascertain the precise scope of each claim involved in the present appeal, it is necessary to refer to the text of each such claim, in its entirety, as set forth in Appendix A hereto.

The present invention relates to a lithium secondary battery in which deterioration of battery properties caused by water released from positive and negative electrodes into a non-aqueous electrolytic solution packed in the battery is suppressed (specification, page 1, lines 5-10).

It is common that water, though the amount is very slight, exists as a contaminant already in the production stage in an organic solvent to be used as a raw material of a non-aqueous electrolytic solution (specification, page 2, lines 17-20). Further, since various kinds of materials and parts constituting a battery, for instance an electrode active material powder, a current collector substrate (a metal foil), a metal terminal, a battery case, and so forth are in general stored in the atmosphere, the moisture adsorbed on the surfaces of these materials and parts may sometimes be taken in the non-aqueous electrolytic solution on the completion of the assembly of a battery (specification, page 2, line 20 - page 3, line 2).

If such water exists in the non-aqueous electrolytic solution, the probability of hydrolysis of the electrolyte is increased and the risk of evolution of an acidic substance, a gas, and the like is heightened, causing undesirable results, including deterioration of the charge-discharge cycle properties (battery capacity alteration characteristic caused by repeated charging and discharging

and hereinafter referred to as cycle properties) and short battery life (specification, page 3, lines 3-11).

Among the causes of taking water in the non-aqueous electrolytic solution is water released from electrode plates after the electrode plates are packed in a battery (specification, page 3, lines 12-14). Though electrode plates are manufactured in strict environments, regarding a battery with a large capacity and produced by winding or laminating electrode plates manufactured by applying electrode active materials to current collector substrates, the moisture adsorbed in the electrode active materials or the like is difficult to completely remove merely by a normal drying process (specification, page 3, lines 15-21; page 4, lines 12-15).

Water contained in an electrode plate and hence existing in the non-aqueous electrolytic solution greatly affects the battery properties and makes it difficult to provide excellent battery properties merely by controlling the water in the non-aqueous electrolytic solution used to fill the battery (specification, page 4, lines 22-27).

The present inventors found that excellent charging and discharging efficiency and cycle properties can be obtained where water released from the positive and negative electrodes after both electrode plates are packed in a battery and consequently existing in a non-aqueous electrolytic solution is at a prescribed value or lower (specification, page 5, lines 3-11).

According to the present invention, there is provided a lithium secondary battery comprising an electrode unit produced by winding or laminating a positive electrode and a negative electrode with a separator therebetween, and a non-aqueous electrolytic solution containing a lithium compound as an electrolyte, wherein the cumulative concentration of water released from both of the positive electrode and the negative electrode in relation to the weight of

the electrode unit, exclusive of current collectors, is suppressed to 5,000 ppm or lower in the case of heating both electrodes at 25 to 200°C and/or to 1,500 ppm or lower in the case of heating the electrodes at 200 to 300°C (specification, page 5, lines 15-26).

The initial charging and discharging efficiency has been verified to be maintained high in cases where water concentration is suppressed to 5,000 ppm or lower when both electrode plates are heated at 25 to 200°C (specification, page 13, line 27 - page 14, line 3). The water released in this temperature range is understood to be water released from the electrode plates in the stage in which the electrode plates are inserted into the battery case and the battery case is filled with the electrolytic solution and the water is understood to affect the initial charging and discharging efficiency (specification, page 14, lines 3-9).

Suitable methods for suppressing the concentration of water released from both electrode plates to 5,000 ppm or lower at the time of heating at 25 to 200°C are (1) slurry preparation in the environment of a low humidity (<30% R.H.), (2) optimization of the drying temperature (>150°C) of coated bodies, and (3) rolled body production in the environment of a low humidity (<30% R.H.) (specification, page 14, lines 10-17).

In cases where the cumulative concentration of water released from the positive and negative electrodes in relation to the weight of the electrode unit besides the current collectors is suppressed to 1,500 ppm or lower in the case of heating both electrodes at 200 to 300°C, the self-discharging efficiency and the cycle properties have been verified to be maintained excellent (specification, page 14, lines 18-25). The water released in this temperature range is understood to be water released from the electrode plates due to activation of water by electric reaction during repeated charging and discharging and the water is believed to affect the self-discharge

and cycle properties deterioration in a middle to long period (specification, page 14, line 24 - page 15, line 3). The methods suitable to be employed as the method for suppressing the concentration of released water to 1,500 ppm or lower at the time of heating both electrode plates at 200 to 300°C are similar to the methods identified above as being suitable for suppressing the concentration of released water at the time of heating the electrode plates at 25 to 200°C (specification, page 15, lines 3-10).

The Karl Fischer method is preferably employed for the measurement of the concentration of water released from both of the positive electrode and the negative electrode (specification, page 16, lines 22-25). Such measurement is carried out by putting both electrodes in an electric furnace, increasing the temperature of the electrodes, and sending the released water to a measurement part through a suction pipe (specification, page 1 line 25 - page 17, line 4). In the Karl Fischer method, a non-aqueous electrolytic solution is dissolved or dispersed in methanol and the resulting sample solution is titrated with a Karl Fischer reagent, and the measurement can be carried out by observing the alteration of the color of the reaction solution by titration (specification, page 17, lines 5-15).

Preferably, the lithium compound contained in the electrolyte is lithium hexafluorophosphate (specification, page 6, lines 1-3).

Preferable compounds to be used for the positive electrode active substance include lithium transition metal compound oxides such as lithium manganese oxide (LiMn_2O_4) (specification, page 8, line 26 - page 9, line 2). Where lithium manganese oxide containing lithium and manganese as main components and having a cubic system spinel structure is used as the electrode active material, the internal resistance of the battery can be kept low (specification,

page 6, lines 5-9).

The compounds to be used for the negative electrode active substance include amorphous carbonaceous material powder such as soft carbon and hard carbon, highly graphitized carbonaceous material powder such as synthetic graphite and natural graphite (specification, page 10, lines 19-23). Preferably, a highly graphitized carbon fiber is used as the negative electrode active substance (specification, page 6, lines 9-11).

The present invention is suitably applied to a battery having a capacity of 2 Ah or more (specification, page 6, lines 11-12). A lithium secondary battery of the present invention is suitably used as an electric power source for driving a motor of an electric automobile or a hybrid electric automobile (specification, page 6, lines 13-17).

Examples 1-5, reported in the specification, demonstrate the importance of the present invention. The electrodes of respective Examples 1 to 5 were produced using various electrode constituent members which were so adjusted as to cause differences in the concentrations of water released from the respective electrodes (specification, page 18, lines 12-16). The other members and the testing environments were the same for all of the samples, and the battery members were sufficiently dried immediately prior to the assembly of the respective batteries to eliminate any effects such as water penetration from outside of the batteries due to defective sealing of the batteries (specification, page 18, lines 16-22).

As shown in Table 1, where the cumulative concentration of water released from the positive and negative electrodes (in relation to the weight of an electrode unit besides the current collectors) was suppressed to 5,000 ppm or lower where the electrodes were heated at 25 to 200°C, it was found that the initial charging and discharging efficiency was kept high

(specification, page 21, lines 15-21). Also, where the cumulative concentration was suppressed to 1,500 ppm or lower where the electrodes were heated at 200 to 300°C, the self-discharging efficiency and cycle properties were kept preferable (specification, page 21, lines 22-25). Batteries having electrode plates satisfying both of the above-described two conditions were found to exhibit excellent initial charging and discharging efficiency, self-discharging efficiency and cycle properties (specification, page 21, line 25 - page 22, line 3).

VI. GROUNDS OF REJECTION

Claims 1-17 are rejected under 35 U.S.C. 103(a) over U.S. Patent No. 6,350,544 (Takami '544) in view of U.S. Patent No. 6,083,644 (Watanabe '644).

VII. ARGUMENT

U.S. Patent No. 6,350,544 (Takami '544)

Takami '544 is directed to a lithium secondary battery having, according to the patent, an improved negative electrode containing a carbonaceous material (Takami '544, col. 1, lines 9-12).

According to Takami '544, lithium secondary batteries have not been put into practical use, mainly because the charge/discharge efficiency of the battery is low and the number of charge/discharge times (or cycle life) thereof is still insufficient (Takami '544, col. 1, lines 44-48). The cause for this poor performance, Takami '544 discloses, is assumed to be ascribed to the fact that lithium constituting the negative electrode is degraded due to a reaction with a nonaqueous electrolyte (Takami '544, col. 1, lines 48-51).

Takami '544 discloses a lithium secondary battery comprising a positive electrode, a negative electrode comprising a carbonaceous material which is capable of absorbing and desorbing lithium ions, and a nonaqueous electrolyte (Takami '544, col. 2, lines 55-59). The carbonaceous material has a region of amorphous carbon structure and a region of graphite structure, and the carbonaceous material has a true density of 1.8 g/cm³ or more and a peak in powder X-ray diffraction which corresponds to not more than 0.340 nm in an interplanar spacing d_{002} derived from (002) reflection (Takami '544, col. 2, lines 60-65).

According to Takami '544, as for the positive electrode active material, various kinds of oxides, such as manganese dioxide, lithium/manganese composite oxide, lithium-containing nickel oxide, lithium-containing cobalt oxide, lithium-containing nickel cobalt oxide, lithium-containing iron oxide and lithium-containing vanadium oxide; or chalcogen compounds such as titanium disulfate or molybdenum disulfate may be employed. Among them, lithium cobalt oxide {for example, $Li_x CoO_2$ (0.8 ≤ x ≤ 1)}, lithium nickel oxide (for example, $LiNiO_2$) and lithium manganese oxide (for example, $LiMn_2O_4$, $LiMnO_2$) are preferable in view of obtaining a high voltage (Takami '544, col. 4, lines 39-49).

U.S. Patent No. 6,083,644 (Watanabe '644)

Watanabe '644 discloses a non-aqueous electrolyte secondary battery which uses, as the negative electrode active material, lithium-containing silicon oxide represented by the compositional formula Li_xSiO_y and uses, as the positive electrode, lithium-containing titanium oxide represented by the general formula $Li_xTi_yO_4$ or lithium-containing iron sulfide represented by the general formula Li_xFeS_y , wherein the values for x and y in each formula are within

specified ranges, so that, according to Watanabe '644, a secondary battery of about 1.5 V having high energy density, less inner resistance and excellent charge and discharge characteristics can be obtained (Watanabe '644, Abstract).

FIG. 1 is a cross-sectional view showing one example of a test cell used for evaluating charge and discharge characteristics of an active material of a non-aqueous electrolyte secondary battery according to Watanabe '644. In the drawing, numeral 1 is a counter electrode case simultaneously used as a counter electrode terminal and is comprised of a plate made of stainless steel, whose outer side surface is Ni-plated; 3 is a counter electrode which is produced such that a predetermined thickness of lithium foil is punched with a diameter of 14 mm and fixed on the inner side surface of the counter electrode case 1; 7 is a working electrode case made of stainless steel whose outer side surface is Ni-plated, and which is also used as a working electrode terminal; 5 is a working electrode formed of an active material according to the invention; 6 is a working electrode collector made of a conductive adhesive using carbon as a conductive filler, and is electrically connected with the working electrode 5 and the working electrode case 7 each of which is adhered together; 4 is a separator made of a porous film of a polypropylene and is impregnated with an electrolyte solution (Watanabe '644, col. 9, lines 9-31).

According to Watanabe '644, “[t]he support salt which can be used is lithium salts [sic] (electrolyte)” such as compounds selected from a list which includes lithium phosphorus hexafluoride (LiPF_6) (Watanabe '644, col. 12, lines 44-51).

According to Watanabe '644, it is desirable that the battery be assembled in a moisture-free atmosphere or an inert gas atmosphere (Watanabe '644, col. 14, lines 37-39). Further, according to Watanabe '644, it is preferable that parts before assembling are previously

dried (Watanabe '644, col. 14, lines 39-41). Watanabe '644 discloses that as drying or moisture-eliminating method of pellet, sheet and other parts, the methods generally used can be employed (Watanabe '644, col. 14, lines 41-43). In particular, according to Watanabe '644, it is preferred to use hot air, vacuum, infrared ray, far infrared ray, electrom [sic] beam, and low moisture air, alone or in combination of two or more thereof (Watanabe '644, col. 14, lines 43-46). According to Watanabe '644, the temperature is preferably in the range of 80 to 350 degrees C, and more preferably 100 to 250 degrees C (Watanabe '644, col. 14, lines 46-48). Watanabe '644 discloses that the moisture content is preferably 2,000 ppm or less as the entire battery, and it is preferred for the positive electrode mixture, the negative electrode mixture or the electrolyte to be 50 ppm or less from the point of cycle property (Watanabe '644, col. 14, lines 48-52).

Comparison of the Claimed Subject Matter With What Would Have Been
Obvious in View of the Applied References

As noted above, Takami '544 discloses that lithium secondary batteries suffer from low charge/discharge efficiency and low cycle life) because lithium constituting the negative electrode is degraded due to a reaction with a nonaqueous electrolyte (Takami '544, col. 1, lines 44-51). Takami '544 is directed to a lithium secondary battery having, according to the patent, an improved negative electrode containing a carbonaceous material (Takami '544, col. 1, lines 9-12).

The negative electrode of Takami '544 comprises a carbonaceous material which is capable of absorbing and desorbing lithium ions (Takami '544, col. 2, lines 55-59). The

carbonaceous material has a region of amorphous carbon structure and a region of graphite structure, and the carbonaceous material has a true density of 1.8 g/cm³ or more and a peak in powder X-ray diffraction which corresponds to not more than 0.340 nm in an interplanar spacing d_{002} derived from (002) reflection (Takami '544, col. 2, lines 60-65).

The positive electrode of Takami '544 can be any of various kinds of oxides, such as manganese dioxide, lithium/manganese composite oxide, lithium-containing nickel oxide, lithium-containing cobalt oxide, lithium-containing nickel cobalt oxide, lithium-containing iron oxide and lithium-containing vanadium oxide; or chalcogen compounds such as titanium disulfate or molybdenum disulfate. Among them, lithium cobalt oxide {for example, $Li_x CoO_2$ ($0.8 \leq x \leq 1$)}, lithium nickel oxide (for example, $LiNiO_2$) and lithium manganese oxide (for example, $LiMn_2O_4$, $LiMnO_2$) are preferable in view of obtaining a high voltage (Takami '544, col. 4, lines 39-49).

The January 14, 2005 Office Action contains an acknowledgment that Takami '544 does not contain disclosure as to the recitation in claims 1 and 17 relating to the water content of the electrodes, namely, that:

a cumulative concentration of water (H_2O) released from both of said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of weight of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C.

As noted above, Watanabe '644 discloses a non-aqueous electrolyte secondary battery which uses $Li_x SiO_y$ as the negative electrode active material and $Li_x Ti_y O_4$ or $Li_x FeS_y$ as the positive electrode active material (Watanabe '644, Abstract).

Watanabe '644 contains no suggestion that any of the features disclosed therein which relate to avoidance of moisture would provide any benefit in batteries which do not employ positive and negative active materials as disclosed in Watanabe '644. Watanabe '644 discloses that it is desirable that "*the battery of the present invention*" be assembled in a moisture-free atmosphere or an inert gas atmosphere. Watanabe '644 contains no suggestion that any benefit would be produced by following the disclosure in Watanabe '644 unless the electrodes of the battery correspond to those described in Watanabe '644, namely, Li_xSiO_y as the negative electrode active material and $\text{Li}_x\text{Ti}_y\text{O}_4$ or Li_xFeS_y as the positive electrode active material.

Any assertion that Watanabe '644 suggests reducing moisture content in *all* lithium secondary batteries, regardless of the electrode active materials, would clearly go beyond what is disclosed in Watanabe '644.

The materials disclosed in Watanabe '644 for use as the positive and negative electrodes do not include any materials disclosed in Takami '544 for use as the positive and negative electrodes of Takami '544.

In addition, neither of the applied references contains any indication that the battery of Takami '544 could be made to have the water content properties disclosed in Watanabe '644, unless the battery includes a negative electrode active material of lithium-containing silicon oxide represented by the compositional formula Li_xSiO_y and a positive electrode active material of lithium-containing titanium oxide represented by the general formula $\text{Li}_x\text{Ti}_y\text{O}_4$ or lithium-containing iron sulfide represented by the general formula Li_xFeS_y .

In addition, the lithium metal oxide material disclosed in Watanabe '644 for use in making the negative electrode can readily absorb water, unlike the carbonaceous material which

the negative electrode active substance recited in claim 1 comprises. Watanabe discloses that the drying or moisture-eliminating temperature is preferably in the range of 80 to 350 degrees C, and more preferably 100 to 250 degrees C. Watanabe further discloses that the battery is assembled after drying in the above ranges. Heating to more than 200 degrees C is not realistic in a case where a battery is assembled after heating, because binder contained in the electrode would normally be decomposed or would deteriorate at such temperatures.

In view of the above, it is respectfully requested that the U.S. PTO reconsider and withdraw this rejection.

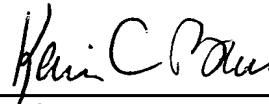
VIII. CONCLUSION

Accordingly, the Honorable Board of Patent Appeals and Interferences is respectfully requested to reverse the Final Rejection of claims 1-17 and to pass this application to allowance and issuance.

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,

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APPENDIX A

1. A lithium secondary battery comprising:
an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator, said positive electrode comprising positive electrode active substance comprising lithium manganese oxide, said negative electrode comprising negative electrode active substance comprising at least one material selected from the group consisting of amorphous carbonaceous material and graphitized carbonaceous material, and
a non-aqueous electrolytic solution containing a lithium compound as an electrolyte, wherein a cumulative concentration of water (H_2O) released from both of said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of weight of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C.
2. The lithium secondary battery according to Claim 1, wherein said lithium compound is lithium hexafluorophosphate.
3. The lithium secondary battery according to Claim 1, wherein a lithium manganese oxide containing lithium and manganese as main components and having a cubic system spinel structure is used as the positive electrode active substance.
4. The lithium secondary battery according to Claim 2, wherein a lithium manganese oxide containing lithium and manganese as main components and having a cubic system spinel structure is used as the positive electrode active substance.
5. The lithium secondary battery according to Claim 1, wherein a graphitized carbon fiber is used as the negative electrode active substance.
6. The lithium secondary battery according to Claim 2, wherein a graphitized carbon fiber is used as the negative electrode active substance.

7. The lithium secondary battery according to Claim 3, wherein a graphitized carbon fiber is used as the negative electrode active substance.
8. The lithium secondary battery according to Claim 1, which has a battery capacity of 2 Ah or more.
9. The lithium secondary battery according to Claim 2, which has a battery capacity of 2 Ah or more.
10. The lithium secondary battery according to Claim 3, which has a battery capacity of 2 Ah or more.
11. The lithium secondary battery according to Claim 4, which has a battery capacity of 2 Ah or more.
12. The lithium secondary battery according to Claim 1, which is used in an electric automobile or a hybrid electric automobile.
13. The lithium secondary battery according to Claim 2, which is used in an electric automobile or a hybrid electric automobile.
14. The lithium secondary battery according to Claim 3, which is used in an electric automobile or a hybrid electric automobile.
15. The lithium secondary battery according to Claim 4, which is used in an electric automobile or a hybrid electric automobile.
16. The lithium secondary battery according to Claim 5, which is used in an electric automobile or a hybrid electric automobile.

17. A lithium secondary battery comprising:

an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator, said positive electrode comprising positive electrode active substance comprising lithium manganese oxide, said negative electrode comprising negative electrode active substance comprising at least one material selected from the group consisting of amorphous carbonaceous material and graphitized carbonaceous material, and

a non-aqueous electrolytic solution containing a lithium compound as an electrolyte, wherein a cumulative concentration of water (H_2O), determined using Karl Fischer's method, released from both of said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of weight of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C.